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PROJECT OFFICERS REPORT—PROJECT 2.13

**RADIOISOTOPE FRACTIONATION AND PARTICLE
SIZE CHARACTERISTICS OF A LOW-YIELD
SURFACE NUCLEAR DETONATION (U)**

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ABSTRACT

Aircraft sampling penetrations of a low-yield nuclear cloud from a land-surface burst in Nevada were made at four altitudes, from 20 to 54 minutes after detonation. Samples from each of the four levels were radiochemically analyzed for 15 fission product isotopes. Other samples from each level were fractionated into seven particle size groups by settling in benzene. The individual size fractions were analyzed radiochemically and by gamma spectrometry. Particle size distributions and specific activities were measured in the untreated and artificially fractionated specimens of debris. Fallout samples were collected in trays along the fallout hot-line. The fallout samples were radiochemically and gamma spectrometrically analyzed.

The extensive radiochemical and physical data published by NRDL on the fallout samples (Reference 9) were used in conjunction with the cloud data to establish: (1) approximately 70 to 75 percent of Zr^{95} and the rare earth refractory fission products had fallen out of the cloud within 20 minutes of the detonation. Of the 25 percent remaining, about 15 percent later fell out locally and at intermediate distances; the remaining 10 percent resided in particles less than 18 micron diameter and was carried to larger distances. (2) Approximately 5 percent of Cs^{137} and Sr^{90} fell out of the cloud in the first 20 minutes. Of the Cs remaining in the 20-minute cloud, about 25 percent fell out locally and at intermediate distances; the remaining 70 to 75 percent was associated with particles less than 18 microns in diameter and was dispersed to larger distances.

The partitioning of 18 fission product chains between the cloud and the prompt fallout has been determined from the detailed radiochemistry of the cloud and fallout samples. All are intermediate between Zr and Cs in partitioning behavior.

The radiochemical composition of prompt fallout particulates can be systematized on the basis of a simple model employing the concept of fallout formation time.

The R value data for the cloud are satisfactorily fit by the relationship $R_{i,cs} = (R_{cs,cs})^n$ where n varies between 0 and 1 for the different isotopes. The relationship is not extrapolatable to the composition of the fallout samples.

Three specific activity and particle size distribution behaviors can be discerned: irregulars in the cloud; irregulars in the prompt fallout; and spheres in the cloud.

The particle frequency functions for the irregulars in the cloud can be fit by $F(D) \propto D^{-2.2}$. The size distribution for spheres appears to be approximately log-normally distributed about a mean of 30 microns.

The specific activity of all isotopes in the cloud is highest in the smallest particles. S_p (fissions of Zr^{95} per gram) in the cloud decreases with increasing particle size but averages about 5×10^{12} , similar to the value in the most

intense prompt fallout samples. S_{137} in the cloud averaged 4×10^{14} fissions/gm compared to 1.2×10^{12} in the intense prompt fallout. The relationship between S_{95} and diameter is given by: $S_{95} \propto D^{-0.2 \pm 0.1}$. S^{137} follows no simple relationship with D over the entire size range but the distribution of Cs between particles $> 18\mu$ and $< 18\mu$ can be fit by $S_{137} \propto D^{-1}$. Data on the regulars (spheres) are tentative. The spheres exhibit a somewhat higher specific activity (S_{95}) than the irregulars. No more than 10 percent of the cloud fission activity is borne by spheres. Microprobe analysis demonstrates most spheres contain iron, with various quantities of Cr, Mn, Ni, and Zn, some of which show a tendency to be enriched relative to iron as particle size increases.

The importance of synthesizing data from cloud and surface sampling analysis programs is stressed. Isotopic fractionation is used as a tool in arriving at a partitioning between prompt and more remote fallout.

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the specific activity of individual airburst particles in the micron size range has been observed by the author to decrease with size. These airburst particles are highly enriched in refractory relative to volatile isotopes, and the majority of their activity is due to the refractory radioisotope components (Zr^{95} and rare earths). Hence one would expect a gross specific activity relationship which is essentially constant, or which falls off between D^2 and D^3 . In fact, the fall off in specific activity with D is much steeper than D^2 in the majority of instances observed (Reference 4). In a surface burst a large fraction of the matrix to which the radioactive components can be attached has never been volatilized at all. Thus surface attachment of the fission fragments to larger inert particles must be considered an important mode of association in ground bursts.

Particle Size Distribution. The NRDL D-Model (Reference 5) for fallout assumes a log-normal particle size distribution for Nevada soil with $\log_{10} D = 2.053$ and $\sigma = 0.732$. Values adopted for coral are $\log_{10} D = 2.209$ and $\sigma = 0.424$. RAND (Reference 5) uses a distribution defined by $\log_{10} D = 1.65$ and $\sigma = 0.69$ to describe 95% of the cloud, and $\log_{10} D = 2.34$ and $\sigma = 0.50$ to describe the remaining 5%. Both models

assume a uniform activity distribution with mass. For example, the RAND model with $\log_{10} D = 1.95$ puts 95% of the activity between 20 and 360 micron diameter particles, and with $\log_{10} D = 2.54$, 95% of the activity is between 160 and 1200 microns.

Specific Activity and Particle Size. The literature on particle size distributions, and specific activity as a function of particle size (mostly gross activity data, undifferentiated by nuclide assay), is abundant. This resume will confine itself to Jangle S, Jangle U, and Teapot ESS (References 6 through 8).

(1) The highest specific activities we have been able to find documented radiochemically for Jangle S and Jangle U are 2.9×10^{13} fissions of Mo^{99} per gram of fallout. In Jangle U, Mo^{99} appeared to be the most refractory isotope, judging from comparative analyses of Zr^{95} and Ce^{144} in the specimens (References 7 and 8). In Teapot ESS, S^{144} (expressed in fissions of Ce^{144} per gram of fallout and estimated by us from the dpm/ga data) exhibited a maximum of about 2.3×10^{13} , along the hot-line, although one sample collected 3,400 yards from ground zero gave an estimated S_{144} of 5×10^{14} fissions per gram (Reference 6). This is probably a sample of true fireball material. It is estimated that one ton of yield will vaporize about 0.4 ton of soil. Thus, a reasonable upper limit for

the specific activity of a soil-like fission product as Ce^{144} would be about 3.5×10^{14} fissions/ μm of prompt fallout. The average specific activity for a given event is difficult to determine without a comprehensive analysis of all pertinent data. For example, in Teapot ESS, S_{144} increased from 4.5×10^{12} at 300 yards (R/hr = 4150) to 2.3×10^{13} at 3,400 yards (R/hr = 43). A crudely weighted average of about 5.5×10^{12} F_{144}/gm is indicated.

(2) The specific activity generally decreased with particle size but was constant to within a factor of two over the range of 400 to 3000 microns. In Jangle S and Jangle U, 95% of the total activity was found in particles in excess of 20 microns diameter. There is some tendency for enrichment of Sr^{89} and Ba^{140} in the finer particles relative to Zr^{95} or Ce^{144} . Typically, ground fallout particles contain less than 10% of their representative Sr^{89} content and about 35% of their representative Ba^{140} content relative to the refractory isotopes (Reference 6 through 8).

(3) The fraction of fully active particle in the intense, prompt fallout increases with particle size. It is estimated in Jangle U that 0.1%, 1%, and 10% of 1, 10, and 100-micron particles, respectively, were active. In Teapot ESS, 12% of particles larger than 200 microns were fully active and 39% were surface active.

(4) Hot particles have a number mean diameter larger than cold particles. The standard deviation of the frequency curves of hot and cold particles are comparable. The mass distribution for the prompt fallout along the hot-line generally is peaked to larger sizes than the undisturbed preshot soil.

(5) The radiochemical composition of cloud and fallout samples indicates severe fractionation of isotopic ratios of a complementary nature. The refractory isotopes Zr^{95} , Ce^{144} , and Mo^{99} are enriched in the fallout samples. The volatile isotopes Sr^{89} , Ba^{140} , Ag^{111} , and Ru^{106} are enriched in the cloud samples. To our knowledge material balance calculations have not been performed for these events, whereby an attempt was made to partition radioisotope activities between surface and the cloud.

TABLE 3.12 RADIOCHEMICAL COMPOSITION OF PROMPT FALLOUT

	<u>Zr⁹⁵ R' Values</u>				
	<u>0-6m</u>	<u>6-20m</u>		<u>0-6m</u>	<u>6-20m</u>
Sr ⁸⁹	0.0317	0.100	I ¹³¹	0.083	0.95
Sr ⁹⁰	0.107	0.40	Te ¹³²	0.061	0.59
Y ⁹¹	0.512	0.82	*Cs ¹³⁶	0.49	-
Mo ⁹⁹	1.00	1.00	Cs ¹³⁷	0.034	0.35
Zr ⁹⁵	1.00	1.00	Ba ¹⁴⁰	0.28	0.67
Ru ¹⁰³	0.74	0.95	Ce ¹⁴¹	0.46 _{+0.15}	0.83
*Ru ¹⁰⁶	0.08	0.40	Ce ¹⁴⁴	1.06	1.00

In the above table, asterisked values have been divided by 1.6 to normalize for their increased yield in bomb spectrum fission. The values in the table for $Sr^{89}/Zr^{95} = .0317$ are estimated to be good to 10% for Sr^{89} , Sr^{90} , Y^{91} , Mo^{99} , Zr^{95} , Ba^{140} , and Ce^{144} and to $\pm 25\%$ of the quoted value for the remaining isotopes, unless noted. The values for $Sr^{89}/Zr^{95} = 0.1$ are considerably less well established and are based exclusively on the lines drawn through the data points of Figures 3.6a and 3.6b.

4.5 PARTICLE SIZE AND MASS FREQUENCY CURVES FOR THE EVENT

The salient features of the particle size and mass frequency curves for the event can be deduced from a synthesis of NRDL and our data. A crude normalization might be provided by using the fact that there are approximately 400 tons of debris in the 20-minute cloud and of the order of 1,000 tons of high specific activity prompt fallout.

The particle size distribution found in the 20-minute cloud appears to be given by $K/D^{+3.25}$ to good approximation between 1 micron and the cloud cut-offs (100 to 160 microns). This implies a mass distribution as $D^{-0.25}$ in the cloud. The NRDL data (see summary in Table 3.11) exhibit a median mass diameter of 700 to 1,000 microns in the more intense fallout (it is lower elsewhere) and a median activity diameter (MAD) of 200 to 300 microns. In addition, it was observed that the fraction of radioactive particles increases with size in the fallout, being about 25 to 50 percent in the larger size fractions (>200 μ). This finding is in accord with our observation that the mean specific activity of the prompt fallout $S_{95} = 5.7 \times 10^{13}$, whereas the specific activity of the lava particles, which represent the main active component, is 1.88×10^{14} or approximately three times higher. Further, the specific activity of lava particles is reasonably constant.

Appendix A
CLOUD PENETRATION
PROCEDURE

Cloud penetration samples were collected by three B-57 sampling aircraft for this project. The aircraft penetrated the Johnie Boy cloud at different altitudes and times after detonation (Table A.1).

Although some modifications of the procedures were made during the experiment just prior to the penetrations, the general procedure for the collection of samples by the aircraft were the same. An aircraft was to penetrate the cloud at a designated altitude and time after detonation in straight and level flight and collect a sample in one of the wing tip sample collectors. After emerging from the cloud, the aircraft would again penetrate at another altitude and collect a sample with the other wing tip sample collector.

The crew was furnished a data card for recording time of entry and exit of the cloud, maximum dose rate recorded in the cloud, total dose during penetration, airspeed, altitude, temperature, and general comments on the size and shape of the cloud.

Immediately after a cloud penetration, the aircraft landed and the crew members were removed from the aircraft and monitored by a Rad Safe team. The aircraft was monitored and the wing tip samples were then removed and placed in individual lead pigs.

DISCUSSION

The Johnie Boy event was detonated one foot below ground at the Nevada Test Site on 11 Jul 62 at 0945 hours. The cloud reached a height of approximately 14,000 feet MSL and ground zero was 5,200 feet MSL.

Three B-57 aircraft from the 1211th Sampling Squadron at Kirtland Air Force Base, New Mexico, performed the cloud penetrations. Two of the aircraft were airborne prior to H-hour and penetrated the cloud from H+20 to H+33 minutes. The third aircraft was dispatched at H+20 minutes and penetrated the cloud at H+48 and H+54 minutes.

The data as recorded by the crew during the cloud penetrations are presented in Table A.2.

Photographs of the cloud at H + 25 seconds, H + 1 minute 30 seconds, and H + 2 minutes 30 seconds, are shown in Figures A.1 through A.3.

After the samples were removed from the aircraft wing tips, they were monitored and placed in lead pigs. The filter sample from a single wing tip tank is divided into a half and two quarter samples. Each of these sections was placed into a single lead pig.

The data obtained from the samples immediately after their removal from the aircraft appear in Table A.3. The data obtained on the samples at H+24 hours appear in Table A.4.

TABLE A.1 DOSE AND DOSE RATE DATA FROM CLOUD PENETRATIONS

MSL Altitude of Penetration (ft)	14,000	11,000	11,000	11,000	9,500	12,000	13,500 rising to 14,000
Penetration Time (min)	H+20	H+20	H+20	H+25	H+33	H+48	H+54
Peak Dose Rate in Cloud (r/hr)	200	60	60	60	6	0.1	8
Average Dose Rate in Cloud (r/hr)	75	10	30	30	4	0.05	4
Time Spent in Cloud (sec)	17	10	15	30	30	68	83
Total Dose (R)	0.6	0.2	0.3	0.45	0.01	0.35	

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